Stimulation of Polariton Photoluminescence in Semiconductor Microcavity

Le Si Dang, D. Heger, R. André, F. Bœuf, and R. Romestain
Laboratoire de Spectrométrie Physique (CNRS UMR 5588), Université J. Fourier—Grenoble, BP 87, F-38402 Saint Martin d'Hères Cedex, France
(Received 27 April 1998)

Photoluminescence at low temperature is studied for a CdTe-based microcavity tuned to resonance with a quantum well exciton. Two distinct stimulation effects are observed with increasing excitation. The first one is associated with the lower polariton state in the strong exciton-photon coupling regime. This effect, whose physical origin has not yet been identified, could be favored by the higher stability of exciton in CdTe. The second stimulation, obtained for much higher excitation, can be assigned to the electron-hole plasma in the weak coupling regime. [S0031-9007(98)07578-4]

PACS numbers: 71.36.+c, 42.55.Sa, 78.45.+h

Optical properties of semiconductor microcavities have attracted new interests since the observation of the strong coupling regime by Weisbuch *et al.* [1]. This regime is realized when the coupling between the exciton confined in quantum wells and the photon mode confined in the cavity exceeds their own damping and lifetime. Experimentally, the strong coupling regime is most easily demonstrated by the anticrossing behavior exhibited by the exciton and the cavity mode when they are brought into resonance. The resulting coupled exciton-photon system is considered as being cavity polaritons in analogy to bulk excitonic polaritons [2]. Basic physical properties of cavity polaritons have been investigated in III-V semiconductor microcavities [3–8], and recently in II-VI semiconductor microcavities as well [9–11]. One of the recent issues is the behavior of cavity polaritons in the nonlinear excitation regime [12–19]. It is well known that excitons could be ionized under high carrier injection, and thus the microcavity system should switch to the weak coupling regime when increasing the carrier density. Indeed, current vertical cavity surface emitting lasers operate by population inversion of the electron-hole plasma in the weak coupling regime. On the other hand, stimulation of polaritons is theoretically expected in any scattering process involving these quasiparticles due to their bosonic character [20]. However, careful spectroscopic measurements in GaAs-based microcavities show no evidence of polariton stimulation since all observed optical nonlinearities with thresholdlike behaviors could be explained in terms of electron-hole correlations in the weak coupling regime [15–17].

As exciton ionization seems to be the limiting factor, II-VI wide bandgap semiconductor microcavities appear to be better candidates for the observation of polariton stimulation. For example, the stability of excitons in ZnSe quantum wells is enhanced due to their smaller Bohr radius, and their active role in stimulated emission is often needed to explain gain measurements [21,22]. Recently photoluminescence (PL) of ZnSe- and CdTe-based microcavities has also been investigated in the nonlinear excitation regime [18,19]. In both studies, the cavity photon mode is at lower energy than the quantum well exciton so that the emitting polariton state is mostly photonlike ($2/3$ photon and $1/3$ exciton in [19]). For high enough excitation a stimulated emission is observed, shifted to the blue with respect to the spontaneous emission by about 2 and 0.3 meV for the ZnSe and CdTe microcavity, respectively. According to the authors, this small shift is evidence that the microcavity is still in the strong coupling regime. This conclusion is of particular interest, not only from the theoretical point of view but also for device application prospects. In this paper, the fundamental issue of polariton stimulation is reexamined under new experimental conditions. Noticing that the usual lasing action in the weak coupling regime of microcavities always takes place at the photonlike mode, it is decided to measure the PL for exact tuning of cavity photon mode and quantum well exciton. This ensures that the polariton states under study are half excitonlike and half photonlike. Moreover, a CdTe-based microcavity optimized for a high splitting-to-linewidth ratio (about 8.5) is used for a better distinction between the strong and weak coupling regimes. Under these experimental conditions, it is unambiguously shown that polariton PL can be stimulated for moderate excitations at low temperature. For higher excitation densities, one observes a shift of the PL line exactly equal to half of the Rabi splitting and the subsequent stimulated emission of the electron-hole plasma in the weak coupling regime.

The sample used in this work is grown by molecular beam epitaxy (MBE) on a $Cd_{0.88}Zn_{0.12}Te$ substrate. It consists of sixteen CdTe quantum wells in a 2λ $Cd_{0.80}Mn_{0.20}Te$ cavity. The quantum wells, 50 Å thick separated by 70 Å of $Cd_{0.80}Mn_{0.20}Te$ barriers, are arranged in three groups of four and two groups of two at the antinodes of the cavity standing wave. The top and bottom cavity mirrors are distributed Bragg reflectors made of thirteen and eighteen pairs of $\lambda/4$ $Cd_{0.75}Mn_{0.25}Te/Cd_{0.40}Mg_{0.60}Te$ layers, respectively. The maximum reflectivity in the stop band is estimated to be about 0.95. The sample is cooled down to 4.2 K for optical measurements. PL is measured by nonresonant excitation into the band-to-band continuum of quantum

 0031-9007/98/81(18)/3920(4)$15.00

wells at about 1.8 eV (above the reflectivity stop band). The excitation source is a dye laser pumped by a frequency doubled Nd:YAG laser that delivers 5 ns pulses at a repetition rate of 10 Hz. In order to obtain a homogeneous excitation spot, the dye laser beam is first expanded and then only a small area in its center part (1:100) is selected and focused to a spot of about 30 μm diameter on the sample. The pulse energy can be varied up to 1 μJ corresponding to a maximum excitation density of about 20 $\mathrm{MW/cm^2}$. Reflectivity and PL measurements are analyzed with a Jobin-Yvon THR-1000 monochromator equipped with a charge-coupled device (CCD) detector.

Figure 1(a) shows the reflectivity spectrum of the microcavity for zero detuning, i.e., when the cavity mode is resonant with the 1 s ground state of the heavy-hole exciton in quantum wells. The upper and lower polariton states are well resolved (FWHM $\approx$ 2.7 meV) at 1681.6 and 1658.7 meV, respectively. The measured Rabi splitting of about 23 meV is consistent with an exciton oscillator strength $f \approx 2.3 \times 10^{13}$ cm^{-2} [10] and an estimated effective number of quantum wells $n_{\mathrm{eff}} \approx 14$. At room temperature, this microcavity is still operating in the strong coupling regime but with a Rabi splitting reduced to about 17 meV because of LO phonon scattering [3]. Figures 1(b) and 1(c) show PL spectra of the microcavity for zero detuning, as a function of the excitation density. For low excitations, the spectrum consists of a single line at 1659.7 meV that can be assigned to the spontaneous emission from the lower polariton. Its width is about the same as in the reflectivity spectrum. No emission from the upper polariton is observed at low temperatures due to thermalization effects. With increasing excitations the PL exhibits successively two thresholdlike behaviors, the first one occurring at 40 $\mathrm{kW/cm^2}$ and the second one at 10 $\mathrm{MW/cm^2}$ (see Fig. 2). The fact that there are 2 orders of magnitude between the two threshold values clearly indicates that their physical mechanisms should be different. Note that PL spectra displayed in Figs. 1(b) and 1(c) are normalized to the corresponding excitation densities. Thus, a superlinear (sublinear) dependence of PL intensity on excitation should give an increase (decrease) of the normalized spectra.

Let us describe first the low threshold nonlinearities. As shown in Fig. 1(b), the lower polariton emission increases linearly with excitations up to about 40 $\mathrm{kW/cm^2}$ since its normalized PL remains unchanged. Then for a further increase of excitation a new line, labeled *A*, emerges from the high-energy shoulder of the lower polariton line (we have carefully checked that similar nonlinearities are obtained also for exciton-photon detunings in the range ± 5 meV). The sharpness of the line (FWHM $\approx$ 0.8 meV as compared to the bare cavity FWHM $\approx$ 4 meV) and its sudden appearance resemble the onset of stimulated emission. However, the most remarkable feature is the spectral position of this line with respect to that expected in the case of transition to the weak coupling regime. For

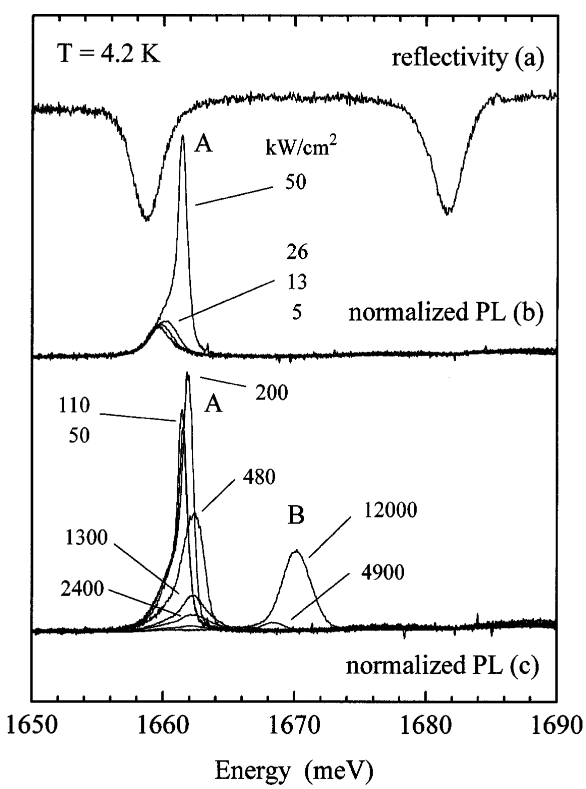

FIG. 1. Optical measurements at 4.2 K and for zero detuning. (a) Reflectivity spectrum. (b) PL spectra for a range of excitation densities. The excitation is at 1.8 eV. All spectra are normalized to the corresponding excitation densities (see text). The *A* line is stimulated PL associated with the lower polariton state in the strong exciton-photon coupling regime. (c) Same as in (b). The *B* line corresponds to the stimulated emission of electron-hole plasma in the weak coupling regime.

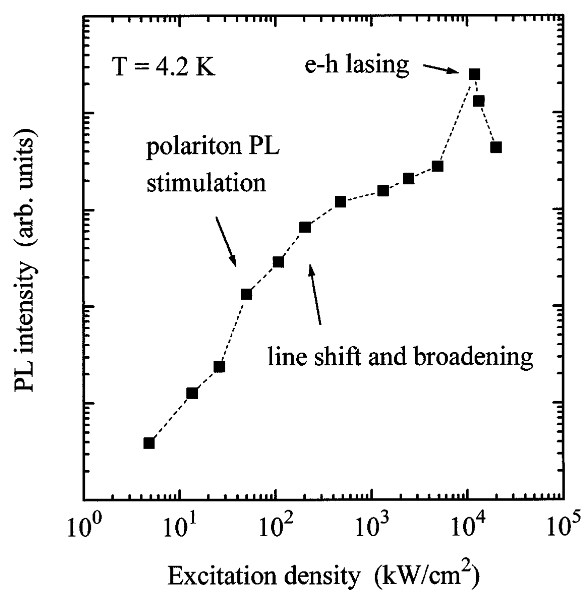

FIG. 2. Integrated PL intensity at 4.2 K, as a function of excitation density. Excitation is at 1.8 eV. Solid squares are experimental data. The dotted line is only a guide for the eyes. Onsets of excitation density effects as observed in Figs. 1(b) and 1(c) are indicated.

zero detuning the bare cavity mode and the bare quantum well exciton are expected at 1670 meV that is well above the position of the *A* line at 1661.4 meV. This clearly shows that the observed PL stimulation actually occurs in the strong exciton-photon coupling regime.

Figure 1(c) shows PL spectra under higher excitation conditions. Starting from 40 $\mathrm{kW/cm^2}$, the *A* emission line increases linearly with excitations up to about 150 $\mathrm{kW/cm^2}$, without any change in its width and in its position. Then for higher excitations up to about 1 $\mathrm{MW/cm^2}$ it broadens continuously and slightly shifts to the blue (by less than 1 meV). For still higher excitations another line, labeled *B*, appears on the high-energy shoulder of the *A* line. It shifts to the blue at the final position at 1670 meV that corresponds exactly to the bare cavity mode (that is also the bare quantum well exciton for zero detuning). Now excitons are completely ionized and the microcavity system is switched to the weak coupling regime. Stimulation at the cavity photon mode (*B* line) is observed for a threshold of about 10 $\mathrm{MW/cm^2}$ (see Fig. 2) and a linewidth FWHM $\approx$ 2.5 meV, that is about half of the bare cavity width. At the highest excitations, the PL intensity eventually decreases, maybe due to heating effects. As in GaAs microcavities [15–17] simulation in the weak coupling regime proceeds via population inversion of the electron-hole plasma. In Fig. 1(c), a detailed examination shows that the *B* line coexists with the *A* line for a certain range of excitations. This is not fully understood for the moment, and the first explanation that comes into mind is a nonuniform distribution of excitation energy over the pump spot. Then the *B* line would be emitted by the part of the pump spot with the highest energy density (probably the spot center) and the *A* line by the part with the lowest density (spot edge).

The experimental data clearly show that the low threshold optical nonlinearities are related to the lower polariton state in the strong exciton-photon coupling regime, confirming the early report in Ref. [19]. They cannot be due to the absorption bleaching phenomenon discussed in [17] because the lower polariton state is not in resonance with any excited state. However, partial saturation of exciton states can result in photon stimulation and bring about conventional lasing if the microcavity losses (mainly mirror losses) are small enough. In CdTe-based microcavities, mirror losses amount to more than 5%. Thus the minimum gain per pass per quantum well needed for conventional lasing would be 0.3% for our 16-quantum-well microcavity and 2.5% for the 2-quantum-well microcavity of Ref. [19]. These gain values are rather high and would imply significant reduction of the exciton oscillator strength, especially for the 2–quantum-well microcavity. If the strong coupling regime is not destroyed yet then one would expect a relative decrease of the Rabi splitting, stronger for the 2-quantum-well microcavity than for the 16-quantum-well microcavity. Actually a blueshift is observed for the lower polariton state in the nonlinear excitation regime: 0.3 meV for the 2-quantum-well microcavity (as compared to 3.8 meV for a complete exciton saturation, Fig. 1 in [19]) and 1.7 meV for the 16-quantum-well one [as compared to 11.5 meV, Fig. 1(b)]. If this blueshift is due solely to a variation of the exciton oscillator strength, it is not consistent with the exciton saturation mechanism. Another possible explanation is stimulation of cavity polaritons as recently proposed by Imamoglu *et al.* [20]. They theoretically show that a strong enhancement of the lowest polariton population can be obtained by relaxation scattering with phonons when the exciton reservoir occupancy exceeds that of the phonons. Since this kind of stimulation effect is a direct consequence of the bosonic character of polaritons, it is to be expected also with other excitonic scatterings, e.g., exciton-exciton interaction.

In any case, the key parameter for stimulation in the strong coupling regime is the degree of exciton stability under high excitation conditions, which is strongly in favor of wide bandgap semiconductors. For example, the saturation density of excitonic resonance (i.e., the density for which the oscillator strength is divided by a factor of 2) is typically $N_s \sim 4.3 \times 10^{10}\ \mathrm{cm^{-2}}$ in 75 Å InGaAs quantum wells [12] and $\sim 6.6 \times 10^{10}\ \mathrm{cm^{-2}}$ in 76 Å GaAs quantum wells [13] for exciton binding energies $E \sim 10$ meV and Bohr radii $a \sim 130$ Å. These values of N_s are probably below the density required for polariton stimulation since no such effect is observed in GaAs-based microcavities [15–17]. In ZnSe quantum wells, excitonic effects are much stronger, yielding binding energies as high as $E \sim 40$ meV [22]. Consequently, the exciton Bohr radius is very small $a \sim 25$ Å which results in an exciton saturation density in the range of $10^{12}\ \mathrm{cm^{-2}}$. Recently, stimulated emission associated with the photonlike mode has been studied in a ZnSe-based microcavity [18]. Although the stimulation mechanism is not clearly identified, gain spectroscopy shows that excitons are not completely ionized at a threshold density of about $2 \times 10^{12}\ \mathrm{cm^{-2}}$. To our knowledge, the saturation density has not been measured in CdTe quantum wells. To have an estimate, we use the theory of Schmitt-Rink *et al.* [23], assuming that the steady electronic state is formed by excitons (see below). Then the saturation density N_s is given by $N_s \pi a^2 \sim 0.117$, where a is a 3D effective Bohr radius which roughly follows the relation $Ea \sim$ const [23]. From exciton bulk values (binding energy $E \sim 10$ meV, Bohr radius $a \sim 70$ Å) and the measured value $E = 25$ meV in our 50 Å quantum wells [10] we deduce $a \sim 28$ Å. This gives a saturation exciton density $N_s \sim 4.8 \times 10^{11}\ \mathrm{cm^{-2}}$, which seems reasonable in regards to GaAs and ZnSe. Unfortunately, the direct comparison of N_s to the exciton density at threshold is made difficult by the use of nonresonant excitation in our measurements. Electron-hole pairs are created by 1.8 eV excitation that is about 140 meV above polariton states. Scatterings between carriers and with (optical) phonons result in fast thermalization and formation of excitons by the bimolecular process. At high excitations, thermalization between carriers and excitons is further accelerated by carrier-exciton scatterings and their relative populations can be estimated from the action mass law [24]. The electronic temperature in our quantum wells is not known, but because of the large exciton binding energy $E = 25$ meV and the high excitation conditions, the exciton population is probably dominant at 4.2 K: For example, an electronic temperature as high as 60 K would yield a population of 90% of excitons after an initial excitation of 10^{10} cm^{-2} electron-hold pairs. Therefore, we assume in the following that only excitons are present after thermalization and relaxation. These excitons with large in-plane wave vectors k are not coupled to the normal cavity mode, and significant radiative losses, as high as 90%, can result from their coupling to leaky photon modes [7]. Finally, a small fraction of the initial pairs will relax to the radiative polariton states ($k = 0$) through the bottleneck region which separates excitons in the weak coupling region from those in the strong coupling region [7,25]. Neglecting nonradiative losses and assuming radiative losses of 90% via leaky modes, the exciton density at threshold is estimated as follows. About half of the excitation intensity goes into the sample and 3% of it is absorbed into each quantum well continuum. Taking 140 ps for the exciton lifetime [26] we obtain an exciton density of about 2.9×10^{10} cm^{-2} at 40 $\mathrm{kW/cm^2}$, which is well below the estimated saturation density $N_s \sim 4.8 \times 10^{11}$ cm^{-2}.

In conclusion, it is unambiguously shown that PL of the lower polariton state can be stimulated at low temperatures in CdTe-based microcavities. This novel effect in the strong exciton-photon coupling regime is confirmed by the observation of transition to the weak coupling regime at much higher excitations. It is made possible by the higher stability of excitons in CdTe quantum wells and should be even more favored in wider bandgap semiconductors such as ZnSe or GaN. However, its physical origin is not identified yet. It could be due to conventional lasing induced by partial saturation of exciton states or to direct stimulation of polariton states via exciton-phonon scattering as discussed by Imamoglu *et al.* [20]. The dependence of stimulation conditions on the temperature, the exciton-photon detuning, and the number of quantum wells in the microcavity is currently being investigated in order to clarify this issue.

The authors thank D. Block, J. Bleuse, and R. Stanley for useful discussions. They are grateful to V. Savona for a critical reading of the manuscript. This work has been completed within the CEA-CNRS joint group "Microstructures de Semiconducteurs II-VI", and is supported by the DSP (DGA Ministère de la Défense).

[1] C. Weisbuch, M. Nishioka, A. Ishikawa, and Y. Arakawa, Phys. Rev. Lett. **69**, 3314 (1992).
[2] V. Savona, F. Tassone, C. Piermarocchi, A. Quattropani, and P. Schwendimann, Phys. Rev. B **53**, 13 051 (1996).
[3] R. Houdré, R.P. Stanley, U. Oesterle, M. Ilegems, and C. Weisbuch, Phys. Rev. B **49**, 16 761 (1994).
[4] R.P. Stanley, R. Houdré, C. Weisbuch, U. Oesterle, and M. Ilegems, Phys. Rev. B **53**, 10 995 (1996).
[5] S. Pau, G. Björk, J. Jacobson, H. Cao, and Y. Yamamoto, Phys. Rev. B **51**, 7090 (1995).
[6] B. Sermage, S. Long, I. Abram, J.Y. Marzin, J. Bloch, R. Planel, and V. Thierry-Mieg, Phys. Rev. B **53**, 16 516 (1996).
[7] J. Bloch and J.Y. Marzin, Phys. Rev. B **56**, 2103 (1997).
[8] D. Whittaker, P. Kinsler, T. Fisher, M.S. Skolnick, A. Armitage, A. Afshar, M. Sturge, and J. Roberts, Phys. Rev. Lett. **77**, 4792 (1996); V. Savona, C. Piermarocchi, A. Quattropani, F. Tassone, and P. Schwendimann, Phys. Rev. Lett. **78**, 4470 (1997).
[9] P. Kelkar, V. Kozlov, H. Jeon, A. Nurmikko, C. Chu, D. Grillo, J. Han, C. Hua, and R. Gunshor, Phys. Rev. B **52**, R5491 (1995).
[10] R. André, D. Heger, Le Si Dang, and Y. Merle d'Aubigné, J. Cryst. Growth **184/185**, 758 (1998).
[11] F. Quochi, G. Hayes, R. André, G. Bongiovanni, A. Mura, J.L. Staehli, and Le Si Dang, J. Cryst. Growth **184/185**, 754 (1998).
[12] R. Houdré, J.L. Gibernon, P. Pellandini, R.P. Stanley, U. Oesterle, C. Weisbuch, J. O'Gorman, B. Roycroft, and M. Ilegems, Phys. Rev. B **52**, 7810 (1995).
[13] J.K. Rhee, D. Citrin, T. Norris, Y. Arakawa, and M. Nishioka, Solid State Commun. **97**, 941 (1996).
[14] S. Pau, H. Cao, J. Jacobson, G. Björk, Y. Yamamoto, and A. Imamoglu, Phys. Rev. A **54**, R1789 (1996).
[15] H. Cao, S. Pau, J. Jacobson, G. Björk, Y. Yamamoto, and A. Imamoglu, Phys. Rev. A **55**, 4632 (1997); R. Huang, H. Cao, and Y. Yamamoto, Phys. Rev. B **56**, 9217 (1997).
[16] F. Jahnke, M. Kira, S.W. Koch, G. Khitrova, E. Lindmark, T. Nelson, Jr., D. Wick, J. Berger, O. Lyngnes, H. Gibbs, and K. Tai, Phys. Rev. Lett. **77**, 5257 (1996).
[17] M. Kira, F. Jahnke, S.W. Koch, J. Berger, D. Wick, T. Nelson, Jr., G. Khitrova, and H. Gibbs, Phys. Rev. Lett. **79**, 5170 (1997).
[18] P. Kelkar, V. Kozlov, A. Nurmikko, C. Chu, J. Han, and R. Gunshor, Phys. Rev. B **56**, 7564 (1997).
[19] J. Bleuse, F. Kany, A. de Boer, P. Chirstianen, R. André, and H. Ulmer-Tuffigo, J. Cryst. Growth **184/185**, 750 (1998).
[20] A. Imamoglu and R. Ram, Phys. Lett. A **214**, 193 (1996); A. Imamoglu, R. Ram, S. Pau, and Y. Yamamoto, Phys. Rev. A **53**, 4250 (1996).
[21] I. Galbraith and S.W. Koch, J. Cryst. Growth **159**, 667 (1996).
[22] J. Ding, M. Hagerott, T. Ishihara, H. Jeon, and A.V. Nurmikko, Phys. Rev. B **47**, 10 528 (1993).
[23] S. Schmitt-Rink, D.S. Chemla, and D.A.B. Miller, Phys. Rev. B **32**, 6601 (1985).
[24] D. Block, R. Romestain, P. Edel, and S. Fränke, J. Lumin. **53**, 339 (1992).
[25] F. Tassone, C. Piermarocchi, V. Savona, A. Quattropani, and P. Schwendimann, Phys. Rev. B **56**, 7554 (1997).
[26] A. Polhmann, R. Hellmann, E. Göbel, D. Yakovlev, W. Ossau, A. Waag, R. Bicknell-Tassius, and G. Landwehr, Appl. Phys. Lett. **61**, 2929 (1992); S. Haacke, N. Pelekanos, H. Mariette, M. Zigone, A. Heberle, and W. Rühle, Phys. Rev. B **47**, 16 643 (1993).